Evidence of illegal emissions of ozone-depleting chemicals

Article

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[Online title:] Evidence of illegal emissions of ozone-depleting chemicals

Chlorofluorocarbons are the main class of chemical that depleted the ozone layer in the stratosphere. Measurements reveal that chlorofluorocarbon emissions are rising again, despite international rules restricting their use. See Letter p.XXX

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Monitoring the expected decline in the atmospheric concentrations of banned compounds might seem like an unexciting research task. But on page XXX, Montzka *et al*.[1] report an unexpected finding in the long-term measurements of CFC-11, one of the most potent ozone-depleting compounds: its atmospheric concentration is decreasing at a much slower pace than would be expected on the basis of its known sources and sinks. This points to a new rise in its emissions — in contravention of international regulations.

CFC-11 belongs to the chlorofluorocarbon (CFC) family of compounds. CFCs are highly stable, synthetic chemicals that were used in a variety of applications since the 1930s — for example as propellants in aerosol sprays, as solvents and as refrigerants in cooling systems. In 1973, the British scientist James Lovelock and his colleagues were the first to measure the abundance of CFCs in the atmosphere and

to realize that these substances were found ubiquitously in both the Northern and Southern Hemispheres, despite their sources being located only in the Northern Hemisphere[2]. This finding triggered the hypothesis that CFCs could be destroyed naturally only in the stratosphere, in a process that releases chlorine atoms. Each of these atoms would be able to destroy many ozone molecules in catalytically driven cycles, thus posing a threat to the ozone layer[3], which protects life on Earth from harmful ultraviolet radiation.

The discovery[4] of the 'hole' in the ozone layer over Antarctica in 1985 proved this hypothesis not only to be correct, but also far more threatening than had been imagined. It spurred research activities to understand why such severe ozone depletion was found over Antarctica alone, and led to political action to restrict the use of CFCs under the Montreal Protocol in 1987. The realization that more-severe ozone depletion would spread further across the globe if CFCs continued to be released into the atmosphere, along with technological advancements that made the replacement of CFCs possible, helped governments to subsequently tighten the regulations on CFCs and ultimately ban their production through several Amendments to the Montreal Protocol. As a result of these actions, CFC concentrations in the atmosphere peaked in the mid-to-late 1990s and have been steadily declining ever since[5]. The Montreal Protocol has been hailed as the most successful international treaty to date that deals with a global environmental issue[6].

Because the destruction of CFCs in the stratosphere is a slow process, their removal from the atmosphere will take many decades. Today's research into stratospheric ozone focuses on whether atmospheric concentrations of ozone-depleting substances are decreasing according to expectations, and whether the ozone layer is on its way to recovery. Working out whether the ozone layer is recovering on the basis of ozone observations alone is particularly difficult, because of confounding effects from natural variability, climate change and ozone pollution[7].

Monitoring the atmospheric concentration of ozone-depleting substances such as CFC-11 is a more direct test of the effectiveness of the Montreal Protocol. However, even for these long-lived chemicals, natural variability in the transport of air masses between the source and sink regions of the chemicals can affect the rate of the expected decline. The source regions are mostly found in highly industrialized areas of the Northern Hemisphere, whereas CFC destruction in the stratosphere (over both hemispheres) acts as the sink. This distribution of sources and sinks leads to concentration differences between the Northern and Southern Hemispheres, which decay in time after emissions cease. The rate of exchange of air between the stratosphere and the underlying troposphere, and between the Northern and Southern hemispheres, both have a crucial role in driving the concentrations of ozone-depleting substances.

Montzka *et al*. have made a rigorous attempt to take into account natural variability in transport between the different regions of the atmosphere to calculate

how it might have generated the observed levels of CFC-11, both by using simple 'box' models and by performing 3D computational simulations using comprehensive climate models that take into account atmospheric chemistry. They conclude that variations in transport alone cannot explain the recent slowing in the rate of decline in CFCs, but that new emissions must have contributed. Additional evidence in support of their hypothesis comes from their observation of an increase in the difference between mean concentrations of atmospheric CFC-11 in the Northern and Southern Hemispheres over the past few years — that is, the excess of CFC-11 in the Northern Hemisphere became larger. Moreover, after 2012, they observed the emergence of a strong relationship between the atmospheric concentration of CFC-11 and the concentrations of other ozone-depleting substances emitted as a result of human activities.

These multiple lines of evidence support their conclusion that changes in atmospheric dynamics, especially in the stratosphere, must be acting in concert with renewed CFC-11 emissions to produce the observed concentrations. Such a careful analysis is crucial, because any claim of renewed — and therefore illegal — emissions will have political implications. Indeed, by taking into account the flow of the atmosphere to the location where the CFC-11 measurements were taken, Montzka *et al*. attribute the renewed emissions to East Asia. They also estimate that these emissions amount to about 13 gigagrams of CFC-11 per year (an increase of 25%) since 2012. However, the uncertainty in the inferred magnitude of new

emissions might be up to 50%, mainly because of the difficulty in working out how air is transported between the stratosphere and the troposphere.

One way to reduce the uncertainties in the estimates and in the probable sources of the renewed emissions would be to use a high-resolution inverse-modelling approach, such as the one that has been promoted[8] in regional studies to attribute sources for emissions of hydrofluorocarbons (the chemicals that replaced CFCs, and which are potential greenhouse gases). However, such an approach would probably need a denser global network of CFC measurements than is currently available. Moreover, the regional inverse models would have to be extended to become global, high-resolution inverse models that include a well-resolved stratosphere and inter-hemispheric transport — which is a tall order, because few such comprehensive modelling systems are currently available that approach the needed resolution.

Montzka and colleagues' study highlights once more that environmental regulations cannot be taken for granted and have to be safeguarded, and that monitoring is required to ensure compliance. Continuous observations of the environment are crucial, not only satellite measurements that yield global coverage, but also from global measurement networks that yield more-accurate *in-situ* data. Taken together with models that encompass both the troposphere and the stratosphere, such data can be used to make defensible inferences about the sources of polluting chemicals.

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[1] Montzka, S., *et al. Nature* **XX**, XXXX (2018).

[2] Lovelock, J. E, *et al. Nature* **241,** doi:10.1038/241194a0 (1973).

[3] Molina, M. J. and F. S. Rowland *Nature,* 249.5460: 8101974.

[4] Farman, J. C *et al. Nature* 6016: 207 (1985).

[5] World Meteorological Organisation (WMO), *Report no 55*, http://esrl.noaa.gov/csd/assessments/ozone/, 416 p. (2014).

[6] Kofi Annan, 2000, presentation at the Millennium Assembly of the United Nations in September 2000 titled "We the peoples: the role of the United Nations in the twenty-first century".

[7] Shepherd, T. G. *et al. Nature Geosci.* **7,** doi:10.1038/NGEO21551 (2014).

[8] Brunner, D. *et al. Atmos. Chem. Phys.* **17,** https://doi.org/10.5194/acp-17-10651-2017 (2017).

Figure 1 | The slowing decline of an ozone-depleting compound. Production of chlorofluorocarbons (CFCs) was banned internationally by the Montreal Protocol in 1987, because CFC emissions lead to the destruction of the stratospheric ozone layer. The blue plot shows the atmospheric abundance[5] of one such compound, CFC-11, in parts per trillion; blue circles indicate past measurements up to 2013, and the

dashed part indicates the projected abundance, assuming that no new CFC-11 is produced. Montzka *et al*.[1] report that the atmospheric abundance of CFC-11 in the past few years (red dots) is greater than had been projected, and so the rate of decline of CFC-11 levels is slower than expected. They conclude that CFC-11 emissions must have increased since 2012.